
(12) **UK Patent Application** (19) **GB** (11) **2 105 247** **A**

(21) Application No **8217925**
(22) Date of filing **21 Jun 1982**
(30) Priority data
(31) **8119373**
(32) **23 Jun 1981**
(33) **United Kingdom (GB)**
(43) Application published
23 Mar 1983
(51) **INT CL³**
B32B 17/04
D02G 3/04
(52) Domestic classification
B5A 1R214E 1R214F
1R314C1A 1R314C1B
1R314C1C 1R314C1S
1R314C6 20T1 C
D1W 7A 7B
U1S 1658 B5A D1W
(56) Documents cited
GB 2088427 A
GB 2077314 A
GB 2039560 A
GB 2017779 A
GB 1535885
GB 1515923
GB 1502843
WO 80/00419 A
(58) Field of search
B5A D1W
(71) Applicants
Courtaulds plc
(Great Britain),
18 Hanover Square,
London W1A 2BB
(72) Inventor
Peter Morgan Griffiths
(74) Agents
J. Y. and G. W. Johnson,
Furnival House,
14-18 High Holborn,
London WC1V 6DE

(54) **Process for making a
fibre-reinforced moulding**

(57) A fibre-reinforced moulding is made by wrapping a core of reinforcing fibres such as carbon and/or aromatic polyamide fibres with a thermoplastic yarn, and thermoforming the wrapped yarn so that the thermoplastic yarn becomes a polymer matrix around the reinforcing fibres. Additional thermoplastic yarn may be included in the core.

The wrapped yarn may be converted into a fabric prior to the thermoforming step, and additional thermoplastic yarns for contributing to the eventual polymer matrix may be incorporated in the fabric, for example as alternate warp or weft yarns in a woven fabric. Unidirectional reinforcement may be achieved by having the wrapped yarn only in the warp or the weft, with thermoplastic yarns comprising the rest of the fabric.

GB 2 105 247 A

SPECIFICATION

Process for making a fibre-reinforced moulding

- 5 This invention relates to a process for making a fibre-reinforced moulding. Component moulded from thermoplastic or thermosetting polymers are often reinforced with fibres where additional strength is required. In the case of thermoplastic
10 polymers this is usually carried out by compounding the polymer with staple fibres in an extrusion compounder. With thermosetting polymers, resin pre-condensate in liquid form is applied to the reinforcing fibres, which may be in a variety of forms includ-
15 ing staple fibres, continuous filaments and woven and non-woven fabrics, and is cured in situ.

- Compounding is an expensive operation requiring care to ensure adequate mixing and is limited to using staple fibres. This limitation does not arise
20 with liquid resin precondensates, but the use of a liquid material and the need for extended curing times impose both practical and economic limitations on the moulding techniques which can be used.

- 25 One proposal which has been made for facilitating blending of reinforcing fibres and matrix polymer is described in British Patent No. 1,200,342, and involves providing the matrix polymer is fibrous form, so that the two types of fibres in staple form
30 are simply blended together on a carding machine. As an alternative, continuous filaments are wound together as a skein.

- A process has now been developed for making a fibre-reinforced moulding in which the blend propor-
35 tions and the distribution of matrix polymer and reinforcing fibre are more accurately controlled, and which allows the reinforcing fibres to be incorporated in the polymer matrix in the form of fabric.

- According to this invention a process for making a
40 fibre-reinforced moulding comprises providing a wrapped yarn having a core component comprising reinforcing fibres, and a wrapping component comprising a yarn of thermoplastic polymer fibres, making an assembly of such wrapped yarn, and heating
45 the assembly under applied pressure to thermoform the wrapping component of the wrapped yarn into a polymer matrix which incorporates said reinforcing fibres.

- The provision of the thermoplastic yarn which is to
50 form a polymer matrix as a wrapping component disposed about the core of reinforcing fibres places the components of the moulding into intimate contact from the start and in a controlled volume ratio. Furthermore, the wrapping component protects the
55 core component of reinforcing fibres, and makes it possible to convert the wrapped yarn into woven fabrics, for example, even when the core component comprises a brittle or substantially untwisted fibre bundle such as a carbon filament tow. The wrapping
60 component deforms thermoplastically during the thermoforming operation to become a polymer matrix, and whilst this is happening the reinforcing fibres of the core component are released from any constraint imposed by the wrapping component and
65 can move under the forces created in thermoform-

ing. In this way, thermoplastic polymer can penetrate between and around the fibres to promote even spreading of fibres within the matrix and improve fibre/matrix adhesion.

- 70 The term 'fibres' is used herein to include both staple fibres and continuous filaments unless the context indicates that staple fibres only are being referred to.

- The core of reinforcing fibres is preferably a yarn
75 or tow of continuous filaments, and the fact that the core is held by a wrapping component allows the yarn or tow to be substantially twistless. This is not only cheaper to produce but allows the filaments to closely approximate the straight condition, which
80 gives maximum tensile strength to the composite structure in that direction and facilitates filament separation during the thermoforming operation. Furthermore, the presence of the wrapping compo-
85 nent makes it unnecessary to size the core yarn or tow for the purpose of its conversion to fabric, which means that there need be no unwanted size material present to interfere with spreading of the reinforcing fibres or their bonding to the matrix polymer.

- Suitable reinforcing fibres include carbon fibres,
90 glass fibres, metal fibres, ceramic fibres, mineral fibres and aromatic polyamide fibres such as "Kevlar" (Registered Trade Mark). Two or more types of reinforcing fibres may be used in combination, for example a yarn or tow of carbon filaments together
95 with a yarn or tow of "Kevlar" (Registered Trade Mark) filaments, to give specific properties such as controlled energy absorption.

- In addition to reinforcing fibres the core compo-
nent may comprise thermoplastic fibres which fuse
100 with the thermoplastic yarn of the wrapping component during subsequent thermoforming of the wrapped yarn and so form part of the polymer matrix. This allows a desired proportion of thermoplastic
105 fibres to be built into the wrapped yarn more easily than if such fibres were confined to the yarn of the wrapping component. This is particularly so where higher proportions of matrix polymer are needed which would otherwise require the use of high count
110 wrapping yarns and/or high wrapping cover and/or multiple wrappings to give the necessary volume of thermoplastic fibre. Any such thermoplastic fibres in the core component should comprise a polymer which fuses compatibly with the polymer of the
115 wrapping component yarn, and both polymers are preferably the same or of the same polymer type.

- The thermoplastic yarn which comprises the
wrapping component may be a continuous filament
yarn, multifilament or monofilament, or a staple
fibre yarn. The polymer from which the yarn is made
120 should be thermoformable at temperatures which do not unduly degrade or melt the reinforcing fibres of the core component. Many thermoplastic polymers are suitable in this regard, including polyamides
125 such as nylon 6, polyesters such as poly(ethylene terephthalate) and poly(butylene terephthalate), polyolefins such as polyethylene, polypropylene and copolymers of ethylene and propylene, polysul-
phones such as polyethersulphone (PES),
polyetheretherketone (PEEK) and polytetra-
130 fluoroethylene (PTFE).

The wrapping component may be applied to the core component in a number of ways. Thus, a spiral wrap, single or multiple, may be produced on a conventional, hollow-spindle wrapping machine, and with this technique a substantially twistless core component may be used. Core-yarn spinning techniques may also be used to produce the wrapped yarn, using folding, doubling or twisting machinery. A newer technique for making core-yarns involves feeding the core component to a machine producing self-twist yarn such as a Repco machine where the yarn components which self-twist about each other envelop the core component in so doing and thereby constitute a wrapping component.

The invention also includes a wrapped yarn suitable for conversion to a fibre-reinforced moulding, said yarn having a core component which comprises fibres selected from carbon fibres, glass fibres, metal fibres, ceramic fibres, mineral fibres and aromatic polyamide fibres, and a wrapping component comprising a yarn of thermoplastic polymer fibres.

The wrapped yarn may be assembled in a variety of ways prior to thermoforming. As yarn, it may be assembled into bundles, warps, arrays or windings. However, for ease of handling and flexibility of design for specific composite structure properties, the wrapped yarn preferably is assembled into the form of a fibrous fabric. The fibrous fabric may be a woven, knitted, braided, non-woven or pile fabric. Accordingly, the invention further includes a fibrous fabric suitable for conversion to a fibre-reinforced moulding, said fabric being constructed from a wrapped yarn according to the invention.

If desired, additional yarns of thermoplastic polymer fibres may be used in conjunction with the wrapped yarn to make the fibrous fabric of the invention. These additional yarns may be woven or knitted into the structure, for example as alternate warps or wefts, or simply laid in at intervals. Another arrangement, which is useful when unidirectional reinforcement is required, is to have one of the warp and the weft of a woven fabric comprising the wrapped yarn of the invention with the other of the warp and the weft comprising the aforesaid additional yarn of thermoplastic polymer fibres. The additional thermoplastic yarns fuse with the thermoplastic fibres of the wrapped yarn during thermoforming and become part of the polymer matrix. The same considerations apply to these yarns as apply to the additional thermoplastic fibres in the core component as outlined earlier.

The thermoforming operation used to convert the thermoplastic wrapping and other yarn into a polymer matrix may be any suitable moulding or forming process, for example pressure moulding, vacuum forming, hot pressing or hot rolling. As an example of one way of making a fibre-reinforced moulding according to the invention, fabric made from the wrapped yarn as described may be cut and stacked in a mould and then subjected to heat and pressure in the mould to convert the thermoplastic fibres to a polymer matrix. Different layers of a stack may be aligned in different directions or comprise fabrics of different construction to give tailored properties to the moulding produced. Thermoplastic

sheet or film may be interleaved in the stack to contribute to the formation of the polymer matrix.

The invention includes a fibre-reinforced moulding produced by the process of the invention. The preferred moulding comprises a thermoplastic polymer matrix reinforced with carbon fibres, preferably in continuous filament form. A percentage by volume of carbon fibres in the moulding (usually known as the volume fraction) of at least 30 usually is required to give good composite properties, with a preferred range being 50 to 65 *per cent*. If the carbon fibres are to be used in conjunction with other reinforcing fibres such as "Kevlar" or glass, then the volume percentage of carbon fibres used may be reduced in proportion to the volume percentage of other reinforcing fibres used.

The invention is illustrated by the following Examples.

Example 1

A substantially untwisted carbon fibre yarn "Grafil" E/XA-S (Registered Trade Mark) of 3,000 filaments and 200 Tex overall count (200 Tex/3K) was passed through the hollow spindle of a wrapping machine with 3 monofilaments (each of 38 Tex) of PEEK, as a combined core component. This was wrapped with a spiral wrapping of a single monofilament of PEEK, also of 38 Tex using a wrapping twist of 473 turns per metre (t.p.m.) in the 'S' mode. The wrapped yarn produced comprised 53 *per cent* by volume carbon filaments and 47 *per cent* by volume PEEK filaments.

The wrapped yarn was woven into a plain weave fabric on a rapier loom at 4.7 ends/cm and 4.7 picks/cm. A stack of 8 layers of this fabric was placed in a mould wherein it was heated to 400°C. under high pressure to form a moulded bar. The bar comprised a matrix of PEEK reinforced by interwoven continuous filaments of carbon aligned longitudinally and transversely of the bar.

Example 2

A wrapped carbon filament yarn was made by the procedure described in Example 1 using a core component comprising a substantially untwisted carbon filament yarn of 200 Tex/3K ("Grafil" E/XA-S) and two ends of a nylon 66 multifilament yarn each of 140 Tex/210 filaments. The wrapping component was a single end of a 44 dTex/10 filament nylon 66 multifilament yarn and the wrapping twist was 473 t.p.m. The wrapped yarn produced comprised 40 *per cent* by volume carbon filaments and 60 *per cent* by volume nylon 66 filaments.

The wrapped yarn was cut into lengths of 150 mm which were laid parallel to the long sides of a mould measuring 150 mm × 10 mm × 2 mm until the mould was filled. After closing of the mould, the wrapped yarns were moulded into a bar using a moulding pressure of 4 GPa and a moulding temperature of 280°C. Moulding was carried out in an atmosphere of nitrogen to minimise degradation of the nylon 66.

Example 3

A substantially untwisted carbon filament yarn "Grafil" E/XA-S (Registered Trade Mark) of 3,000 filaments and 200 Tex overall count (200 Tex/3K) was passed through the hollow spindle of a wrapping

machine together with a glass filament yarn (204 Tex/1224 filament) and seven monofilaments (each of 38 Tex) of PEEK, as a combined core component. This was wrapped with a spiral wrapping of a single monofilament of PEEK, also of 38 Tex, using a wrapping twist of 473 t.p.m. in the 'S' mode. The wrapped yarn produced comprised 53 per cent by volume reinforcing fibres (carbon and glass) and 47 per cent by volume PEEK filaments.

The wrapped yarn was woven into a plain weave fabric on a rapier loom at 4.0 ends/cm and 4.0 picks/cm. A stack of 8 layers of this fabric were then moulded into a bar as described in Example 1.

Example 4

A wrapped yarn was made as in Example 1. This yarn was woven as the warp of a fabric at 4.7 ends/cm on a rapier loom using a 38 Tex PEEK monofilament as weft yarn at 4.7 picks/cm.

The fabric produced was moulded into a bar as described in Example 1, with the carbon fibre yarn aligned longitudinally of the bar and constituting 48 per cent by volume of the moulded bar.

Example 5

A substantially untwisted carbon filament yarn "Grafil" E/XA-S of 200 Tex/3K was passed through the hollow spindle of a wrapping machine together with a 142 Tex/1K filament yarn of poly(paraphenylene terephthalamide) ("Kevlar") and two ends of a nylon 66 multifilament yarn each of 140 Tex/210 filaments. The wrapping component was a single end of a 44 dTex/10 filament nylon 66 multifilament yarn, and the wrapping twist was 473 t.p.m.

The wrapped yarn comprised 32 per cent by volume carbon fibres, 22 per cent by volume "Kevlar" fibres and 46 per cent by volume nylon 66 fibres.

The wrapped yarn was moulded into a bar as described in Example 2.

Example 6

Two substantially untwisted carbon filament yarns "Grafil" E/XA-S of 200 Tex/3K were passed through the hollow spindle of a wrapping machine together with two "Kevlar" yarns each of 142 Tex/1K and five nylon 66 yarns each of 140 Tex/210. The wrapping component was a single end of a 44 dTex/10 nylon 66 multifilament yarn, and the wrapping twist was 473 t.p.m.

The wrapped yarn comprised 26 per cent by volume carbon fibres, 28 per cent by volume "Kevlar" fibres and 46 per cent by volume nylon 66 fibres.

The wrapped yarn was moulded into a bar as described in Example 2.

CLAIMS

1. A process for making a fibre-reinforced moulding comprising providing a wrapped yarn having a core component comprising reinforcing fibres and a wrapping component comprising a yarn of thermoplastic polymer fibres, making an assembly of such wrapped yarn, and heating the assembly under applied pressure to thermoform the wrapping component of the wrapped yarn into a polymer matrix which incorporates said reinforcing fibres.

2. A process as claimed in claim 1 in which the wrapped yarn has a core component comprising thermoplastic polymer fibres in addition to the reinforcing fibres whereby the thermoplastic polymer

fibres of the core component are thermoformed together with the thermoplastic polymer fibres of the wrapping component to form a polymer matrix which incorporates said reinforcing fibres.

3. A process as claimed in claim 2 in which the thermoplastic fibres of the core and wrapping components of the wrapped yarn comprise the same polymer.

4. A process as claimed in any of claims 1 to 3 in which the wrapped yarn has a core component of which the reinforcing fibres comprise one or more substantially twistless continuous filament yarn(s) or tow(s).

5. A process as claimed in any of claims 1 to 4 in which the wrapped yarn has a core component of which the reinforcing fibres comprise fibres selected from carbon fibres, glass fibres, metal fibres, ceramic fibres, mineral fibres and aromatic polyamide fibres.

6. A process as claimed in any of claims 1 to 5 in which the wrapped yarn has a core component comprising reinforcing fibres of two or more types.

7. A process as claimed in any of claims 1 to 6 in which the wrapped yarn has a wrapping component which comprises one or more spirally-wrapped yarns of thermoplastic polymer fibres.

8. A process as claimed in any of claims 1 to 7 in which the wrapped yarn has a wrapping component which comprises one or more continuous filament yarns of thermoplastic polymer fibres.

9. A process as claimed in any of claims 1 to 8 in which the wrapped yarn has a wrapping component which comprises yarn of thermoplastic polymer fibres selected from fibres of thermoplastic polyamides, polyesters, polyolefins, polysulphones, polyetheretherketone, and polytetrafluoroethylene.

10. A process as claimed in any of claims 1 to 9 in which the wrapped yarn is made into an assembly comprising a fibrous fabric.

11. A process as claimed in claim 10 in which the fibrous fabric comprises yarn of thermoplastic polymer fibres in addition to the wrapped yarn whereby said additional yarn is thermoformed together with the yarn of thermoplastic polymer fibres which comprises the wrapping component of the wrapped yarn, and any thermoplastic polymer fibres in the core component of the wrapped yarn, to form a polymer matrix which incorporates said reinforcing fibres.

12. A process as claimed in claim 10 or claim 11 in which the fibrous fabric is a woven or knitted fabric.

13. A process as claimed in claim 12 in which the fibrous fabric is a woven fabric having one of the warp and the weft thereof comprising said wrapped yarn and the other of the warp and the weft comprising yarn of thermoplastic polymer fibres.

14. A process for making a fibre-reinforced moulding substantially as hereinbefore described in any of the Examples.

15. A process for making a fibre-reinforced moulding substantially as hereinbefore described.

16. A fibre-reinforced moulding made by a process as claimed in any of claims 1 to 15.

17. A wrapped yarn suitable for conversion to a

fibre-reinforced moulding, said yarn having a core component which comprises fibres selected from carbon fibres, glass fibres, metal fibres, ceramic fibres, mineral fibres and aromatic polyamide fibres, and a wrapping component comprising a yarn of thermoplastic polymer fibres.

18. A wrapped yarn as claimed in claim 17 in which the core component comprises thermoplastic polymer fibres in addition to the reinforcing fibres.

19. A wrapped yarn as claimed in claim 18 in which the thermoplastic fibres of the core and wrapping components comprise the same polymer.

20. A wrapped yarn as claimed in any of claims 17 to 19 in which the reinforcing fibres of the core component comprise one or more substantially twistless continuous filament yarn(s) or tow(s).

21. A wrapped yarn as claimed in any of claims 17 to 20 in which the reinforcing fibres of the core component comprise two or more types.

22. A wrapped yarn as claimed in any of claims 17 to 21 in which the wrapping component comprises one or more spirally wrapped yarns of thermoplastic polymer fibres.

23. A wrapped yarn as claimed in any of claims 17 to 22 in which the wrapping component comprises one or more continuous filament yarns of thermoplastic polymer fibres.

24. A wrapped yarn as claimed in any of claims 17 to 23 in which the wrapping component comprises yarn of thermoplastic polymer fibres selected from fibres of thermoplastic polyamides, polyesters, polyolefins, polysulphones, polyetheretherketone and polytetrafluoroethylene.

25. A wrapped yarn substantially as hereinbefore described in any of the Examples.

26. A wrapped yarn substantially as hereinbefore described.

27. A fibrous fabric suitable for conversion to a fibre-reinforced moulding, said fabric being constructed from a wrapped yarn as claimed in any of claims 17 to 26.

28. A fibrous fabric as claimed in claim 27 comprising yarn of thermoplastic polymer fibres in addition to the wrapped yarn.

29. A fibrous fabric as claimed in claim 26 or claim 27 which is a woven or knitted fabric.

30. A fibrous fabric as claimed in claim 29 which is a woven fabric having one of the warp and weft thereof comprising said wrapped yarn and the other of the warp and the weft comprising yarn of thermoplastic polymer fibres.

31. A fibrous fabric substantially as hereinbefore described in any of the Examples.

32. A fibrous fabric substantially as hereinbefore described.